

# Anisotropic densification of glass powder compacts

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**ABSTRACT.** Anisotropic densification of glass powder compacts.

The sintering of a non-spherical aluminosilicate glass powder was investigated using a heating microscope. The samples showed shrinkage anisotropy. The densification rate shows a stronger dependence on density than the prediction of Scherer's theory. The ratio of the creep rate due to anisotropic shrinkage to the densification rate shows no significant variation with density but it depends on the degree of shrinkage anisotropy.

**KEY WORDS.** Anisotropy, shrinkage, glass powders.

**RESUMEN.** Densificación anisotrópica de compactos de polvos de vidrio.

Se estudia en este trabajo la sinterización de polvos de vidrio de silicoaluminato no esféricos, mediante microscopía de calefacción. Las muestras presentaron contracción anisotrópica y la velocidad de densificación presenta una dependencia más fuerte con la densidad que la que predice la teoría de Scherer. El cociente entre la velocidad de fluencia debido a la contracción anisotrópica y la velocidad de densificación no muestra una variación significativa con la densidad pero sí depende del grado de contracción anisotrópica.

**PALABRAS CLAVE.** Anisotropía, contracción, glass powders.

## 1. INTRODUCTION

The sintering of glass has been frequently investigated and important results concerning densification and creep during sintering have been obtained. Most of these works were performed by means of dilatometric measurements and by application of low uniaxial stresses (1,2).

Experimental results have been compared with the predictions of theoretical models for viscous sintering, Scherer's model (3) being the one that appears to have a broader applicability than other available models (4) because it assumes a particular geometry that can be applied to the entire densification process.

Earlier experimental work of Rahaman et al. (1) on sintering of crushed soda-lime glass powder provided interesting results that however showed ambiguity by the comparison with the predictions of Scherer's model. For example both the densification rate and the creep viscosity showed a much stronger dependence on density compared with the theoretical predictions. They also found that the shrinkage of the samples was less in the radial than in the axial direction, in contrast to previous investigations on the sintering behaviour of cordierite-type glass powder (5,6) where the ratio of the axial to the radial shrinkage was about 0.7 for samples formed by pressing in the axial direction.

In a recent paper Rahaman and De Jonghe (2) have studied the sintering of a well characterized spherical borosilicate glass powder and they found in this case excellent agreement with Scherer's theory. The direction of the anisotropic shrinkage coincided with the results of Giess et al. (5,6), i.e. the samples shrank more in the radial than in the axial direction. Particle alignment produced during compaction and non uniform binder burnout appear to be the main causes of the unexpected shrinkage anisotropy observed for the crushed soda-lime glass powder (1). In recent paper (2) the necessity of further work in this field was emphasized in order to determine if the strong dependence

of the densification rate on density is a general result for nonspherical glass powders and to investigate the causes of the unexpected shrinkage anisotropy also found earlier.

In this work results for the densification and creep rates during sintering of a non-spherical aluminosilicate alkali-free glass powder are reported. The sintering was performed in a heating microscope, that allowed monitoring of the axial and radial shrinkage without the exertion of any external load. The densification rate obtained from the measurements is compared with the predictions of Scherer's theory. A similar investigation, but dealing with a borosilicate glass powder has been already conducted (7).

## 2. EXPERIMENTAL

The non-spherical glass powder used was a commercially available\* aluminosilicate alkali-free glass with a mean particle size of 8  $\mu\text{m}$  and theoretical density of 2.57  $\text{g/cm}^3$ . The powder was used in the as-received condition and the particle morphology was analyzed using SEM. Green cylindrical compacts (5 mm. in diameter by 5 mm.) were obtained by uniaxial pressing at room temperature without the addition of any binder. Pressures of 25 MPa were used to reach green densities of  $0.54 \pm 0.02$  of the theoretical. Sintering was performed in a heating microscope\* for 2 hours at 850° C. in air without the exertion of any external load. The samples were placed on an alumina plate, their axis coinciding with the vertical. After sintering, the compacts retained their cylindrical shape, so that friction between sample and substrate could be neglected. The furnace was first heated to the sintering temperature and then the compacts were introduced quickly, so that all the sintering process was performed at isothermal conditions. During the sintering process, photographs of the samples were taken, which allowed the measurements of instantaneous lengths and diameters and hence the calculation of axial and radial

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shrinkage. The mass and dimension of green and sintered compacts were measured and the geometrical densities determined. The final density of the sintered compacts was also measured using Archimede's principle. SEM was used to microstructural examination of sintered compacts.

### 3. RESULTS AND DISCUSSION

Fig. 1 shows a SEM micrograph of the glass powder used. The nonspherical character of the particles is evident.

The density of the compacts at given times during sintering was determined from the green density and the measured shrinkage using the relation

$$\rho = \frac{\rho_0}{(1-\Delta R/R_0)^2 (1-\Delta L/L_0)} \quad [1]$$

where  $\Delta R = R_0 - R$  and  $\Delta L = L_0 - L$ .  $R_0$ ,  $L_0$  are the initial radius and length of the sample and  $R$ ,  $L$  are the respective instantaneous values.

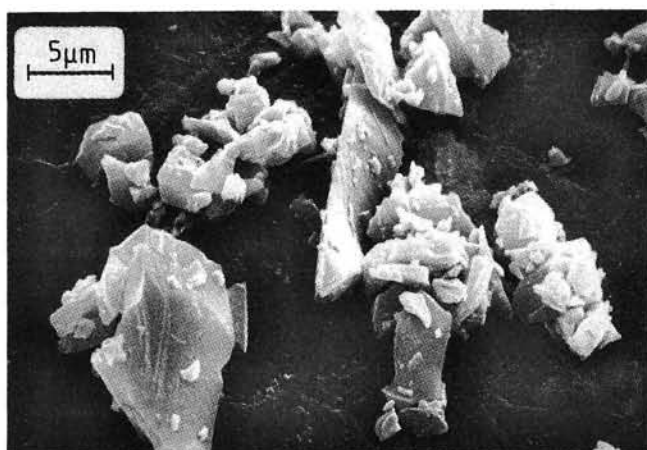


Fig. 1. SEM micrograph of the aluminosilicate alkali-free glass powder used.

The relative density as a function of sintering time is represented in fig. 2. The data shown are an average of two runs under the same conditions and they were reproducible to within  $\pm 4$  %.

From the data for the axial and radial shrinkage, the respective strain rates were calculated, as in earlier works, using the following relations (1,2):

$$\dot{\epsilon}_z = \frac{d[\ln(L/L_0)]}{dt} \quad [2]$$

$$\dot{\epsilon}_r = \frac{d[\ln(R/R_0)]}{dt} \quad [3]$$

Fig. 3 shows the results for axial ( $\epsilon_z$ ) versus radial ( $\epsilon_r$ ) strain, indicating the anisotropic character of the densification. The shrinkage is lower in the axial than in the radial direction as expected. This result coincides with the results for spherical and non-spherical borosilicate glass powder (2,7) and for cordierite-type glass powder (5) but is quite different from that found in the earlier work on crushed soda-lime glass powder (1), in which the samples shrank more in the axial direction.

This result confirms that anisotropy in shrinkage is not only influenced by particle shape but also other factors, such as particle size distribution, particle alignment and process-

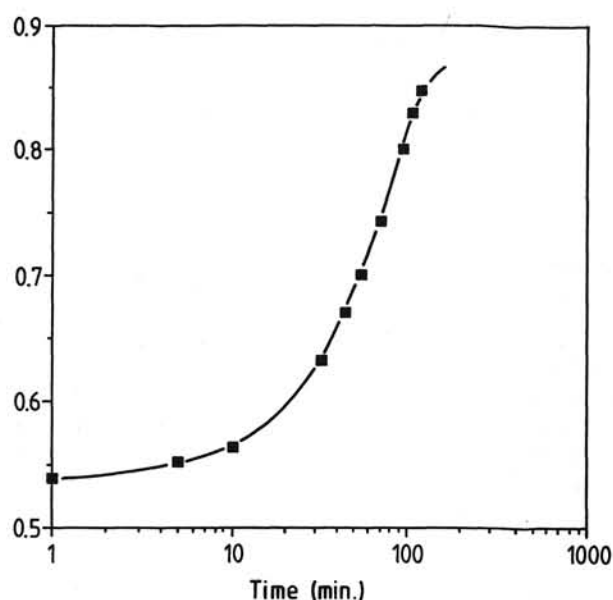


Fig. 2. Relative density versus sintering time for cylindrical glass powder compacts sintered at 850° C under no external load. The axis of the sample coincides with the vertical.

ing conditions (temperature, binder burnout, green density) should be relevant. The slope of the straight line in fig. 3 is the strain anisotropy ratio  $\epsilon_z/\epsilon_r$  and its value is 0.85, a little lower than the shrinkage anisotropy found in previous works (2,5,7). The values for the densification ( $\dot{\epsilon}_p$ ) and creep ( $\dot{\epsilon}_c$ ) rates can be determined according to the formulation introduced by Rak (8) and already used (1,2,7):

$$\dot{\epsilon}_c = \frac{2}{3}(\dot{\epsilon}_z - \dot{\epsilon}_r) \quad [4]$$

$$\dot{\epsilon}_p = \frac{\dot{\rho}}{\rho} = -(\dot{\epsilon}_z + 2\dot{\epsilon}_r) \quad [5]$$

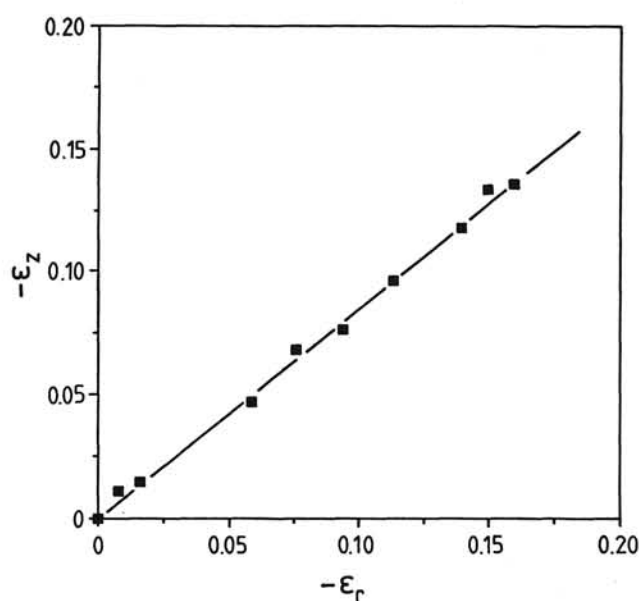


Fig. 3. Axial strain versus radial strain for the conditions described in Figure 2.

Although in the present experiments there are no applied loads, the word «creep» is used to be in agreement with the related literature (1,2).

The experimental results for  $\dot{\epsilon}_p$  and  $\dot{\epsilon}_c$  as a function of density are shown in fig. 4.

Following a suggestion of Rahaman and De Jonghe (2), an objective of this work is to provide experimental data for

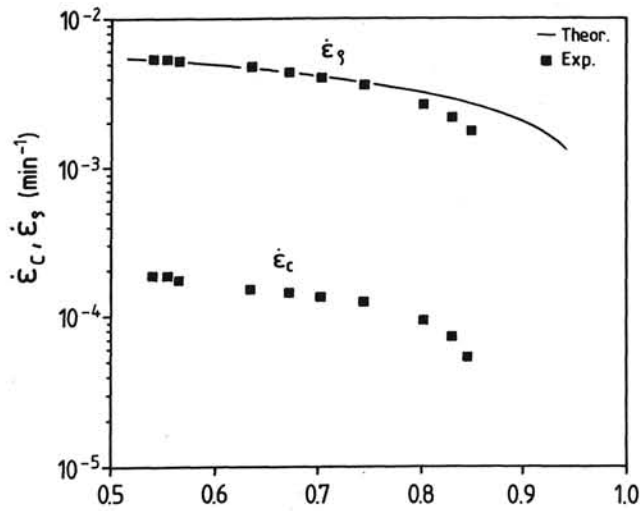


Fig. 4. Creep rate and densification rate obtained from the measurements versus relative density. Also shown are the theoretical values for densification rate from Scherer's model.

the densification rate of the non-spherical borosilicate glass powder as a function of density and to compare these results with the theoretical predictions of Scherer's model. According to this model (3) the densification rate of a compact with isotropic pore size distribution is given by

$$\dot{\epsilon}_p = C \frac{(2 - 3.6 a/l)}{(a/l)^{1/3} (1 - 1.2 a/l)^{2/3}} \quad [6]$$

where C is a constant related to material and geometrical properties,  $a$  and  $l$  are respectively the radius and length of the cylinders of the model.  $a/l$  and the density are related by the equation.

$$\rho = 3 \Pi (a/l)^2 - 8 \sqrt{2} (a/l)^3 \quad [7]$$

In fig. 4 are represented the theoretical results for  $\dot{\epsilon}_p$ . The constant C in Eq. 6 was arbitrarily chosen to give agreement between theory and experiment at the beginning of the sintering process. It is seen that the experimental densification rate shows a stronger variation than predicted by Scherer's theory. As in the previous work (1), this deviation between theory and experimental can be explained considering the differences between the actual pore morphologies and that assumed in the model (3), consisting of cylinders connected into a cubic array. The pore microstructure of a sample sintered to a relative density of 0.8 is shown in fig. 5 indicating the departure of the real pore shapes from the model geometry. The presence of a distribution in pore sizes, as also indicated in fig. 5, may also lead to the deviation

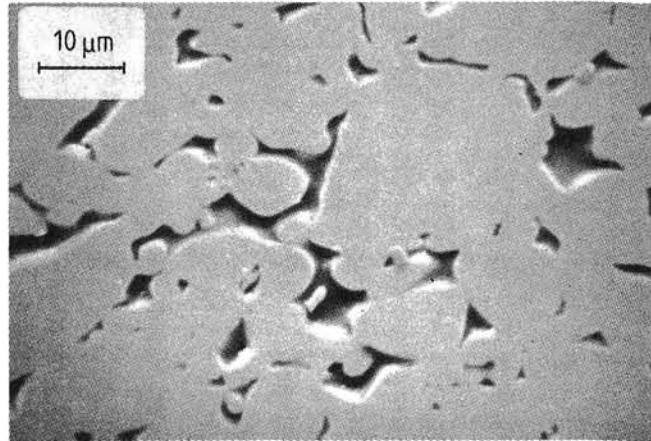


Fig. 5. Pore microstructure of a glass compact sintered to a relative density of 0.8.

between theory and experiment. As shown in (9), the variation of the densification rate depends on the pore size distribution.

An interesting result of these experiments is that the ratio  $\dot{\epsilon}_c/\dot{\epsilon}_p$  is independent of the density as shown in fig. 6. The relation between creep rate and densification rate for this glass powder sintered at 850° C can be written as

$$\dot{\epsilon}_c = 0.035 \dot{\epsilon}_p \quad [8]$$

Rahaman et al. (1) found a value of 0.14 for the ratio  $\dot{\epsilon}_c/\dot{\epsilon}_p$  studying the sintering of crushed soda-lime glass powder at 605° C. If the temperature does not play a role by affecting

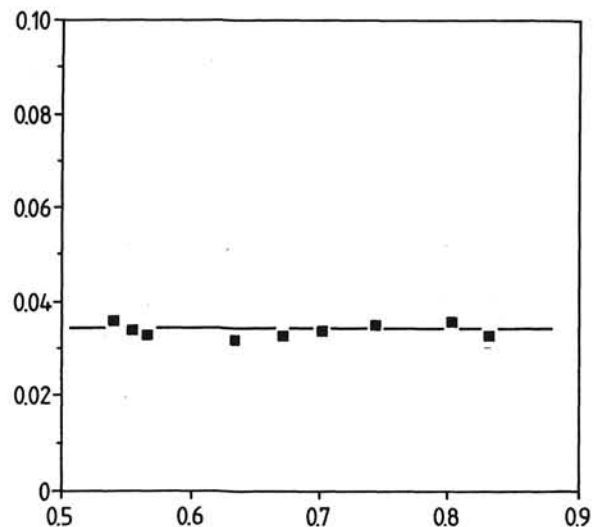


Fig. 6. Ratio of creep rate to densification rate versus relative density.

this ratio because both processes (creep and densification) occur by the same mechanism of viscous flow, the lower creep rate found in this study can be due to a more uniform compact microstructure and therefore lower anisotropy in the shrinkage. Indeed, the ratio  $\epsilon_z/\epsilon_r$  found in this study is 0.85 while in the case of crushed soda-lime glass powder (1) this ratio was 1.8. From Eq. 4 and 5 and considering that the

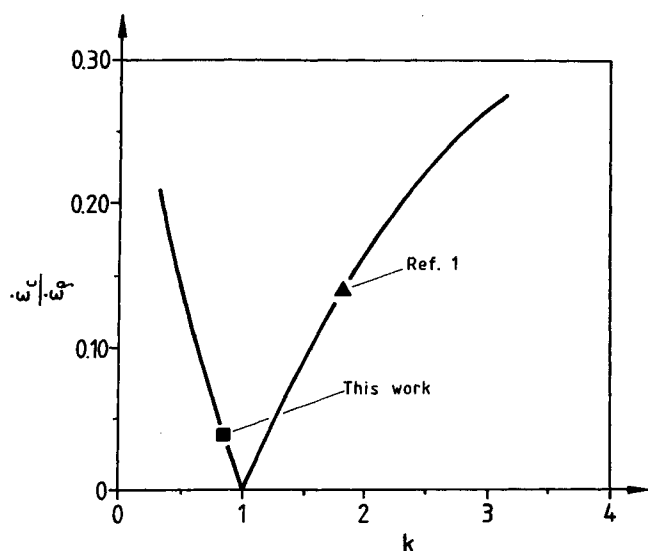


Fig. 7. Variation of the ratio of creep rate to densification rate with the grade of shrinkage anisotropy.

ratio of the axial to the radial shrinkage remains constant during the sintering process ( $\epsilon_z/\epsilon_r = k$ ) it is possible to write

$$\frac{\dot{\epsilon}_c}{\dot{\epsilon}_p} = \frac{2/3(k-1)}{(2+k)} \quad [9]$$

Fig. 7 shows the experimental values for the ratio  $\dot{\epsilon}_c/\dot{\epsilon}_p$  from this study and according to Rahaman et al. (1) plotted versus the respective anisotropy ratio  $k = \epsilon_z/\epsilon_r$ . Also is shown the curve represented by Eq. 9. Thus, it is possible to imagine a way of varying the ratio  $\dot{\epsilon}_c/\dot{\epsilon}_p$  without the exertion of external loads and only with variation of the degree of anisotropy of shrinkage.

#### 4. CONCLUSIONS

Using the heating microscope provides an appropriate experimental method to study the sintering kinetics: data for creep and densification rates are obtained without the exertion of external loads.

The non-spherical aluminosilicate alkali-free glass powder sintered at 850° C showed the expected shrinkage anisotropy, i.e. the samples shrank less in the axial direction, which is the direction of pressing during the formation of green compacts.

The densification rate showed a stronger dependence on density than the prediction of Scherer's model. Differences between actual pore morphologies and that assumed in the model and the presence of a pore size distribution are the reasons for these deviations.

The ratio of the creep rate to the densification rate is 0.035 and it is almost independent of the density. It is interesting to observe that this ratio can be increased by enhancing the degree of anisotropy of shrinkage, which is the unique source of creep when no external loads are exerted during sintering.

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